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(71) 出願人 000003159

東レ株式会社

東京都中央区日本橋室町2丁目2番1号

(72) 発明者 沖田 茂

東京都中央区日本橋室町2丁目2番1号

東レ株式会社内

(72) 発明者 鈴木 篤

愛知県名古屋市港区大江町9番地の1 東

レ株式会社名古屋事業場内

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(54) 【発明の名称】 熱可塑性樹脂組成物及び成形品

(57) 【要約】

【課題】 導電性、ガスバリア性、強度、耐腐食性、成形性に優れ、燃料電池セパレーターに適した熱可塑性樹脂組成物を提供する。

【解決手段】 炭素繊維、カーボンナノチューブを熱可塑性樹脂に配合する。

【0003】燃料電池の各セルは、反応を司る電解質、 50

【0007】このように従来の材料・方法では導電性、ガスバリア性、強度、耐腐食性に優れたセパレーターを効率良く、安価に製造するための材料や方法が見出されていなかった。

【0008】そこで本発明ではこのような用途に適しており、ガスバリア性、強度、耐腐食性、成形性に優れた材料および成形品を提供することを課題とする。

【0009】

【課題を解決するための手段】そこで本発明者らは上記の課題を解決すべく鋭意検討した結果、熱可塑性樹脂に炭素繊維とカーボンナノチューブを併用して配合することにより、熱可塑性樹脂の成形性を損なうことなく高強度化でき、かつ高度な導電性を付与し得ることを見出し、本発明に到達した。

【0010】すなわち、本発明は、

1. 炭素繊維及びカーボンナノチューブを含有する熱可塑性樹脂組成物、
2. 炭素繊維の配合量が10～70重量%、カーボンナノチューブの配合量が0.1～15重量%である前記1記載の熱可塑性樹脂組成物、
3. 炭素繊維の数平均繊維長(L)と繊維径(d)との比(L/d)が10～10,000である前記1または2記載の熱可塑性樹脂組成物、
4. 熱可塑性樹脂がポリオレフィン、ポリアミド、ポリアセタール、非液晶ポリエステル、ポリフェニレンオキシド、ポリカーボネート、ポリフェニレンスルフィド、液晶ポリエステル、ポリエーテルケトン、ポリエーテルエーテルケトン、ポリスルホン、ポリエーテルスルホン、ポリエーテルイミドおよびこれらのブレンド物からなる群から選ばれる一種以上である前記1～3のいずれかに記載の熱可塑性樹脂組成物、
5. 熱可塑性樹脂がポリフェニレンスルフィドまたは液晶ポリエステルである前記1～3のいずれかに記載の熱可塑性樹脂組成物、
6. 熱可塑性樹脂がポリフェニレンスルフィドである前記1～3のいずれかに記載の熱可塑性樹脂組成物、
7. 前記1～6のいずれかに記載の熱可塑性樹脂組成物を成形して得られる成形品、
8. 成形品中の炭素繊維の数平均繊維長(L)と繊維径(d)との比(L/d)が5～5,000である前記7に記載の成形品、
9. 前記1～6のいずれかに記載の熱可塑性樹脂組成物を成形して得られる燃料電池用セパレーター、
10. 成形品中の炭素繊維の数平均繊維長(L)と繊維径(d)との比(L/d)が5～5,000である前記9に記載の燃料電池用セパレーターを提供するものである。

【0011】

【発明の実施の形態】以下に本発明を詳細に説明する。

【0012】本発明で使用される炭素繊維はポリアクリル繊維を原料とするPAN系炭素繊維、石油精製時の残さであるピッチを原料とするピッチ系炭素繊維のいずれも使用できる。

【0013】本発明で使用される炭素繊維の径について

は特に制限はないが、通常1～30μmであり、好ましくは3～20μmであり、更に好ましくは5～15μmである。

【0014】本発明で使用される炭素繊維の長さには特に制限はない。長い方が機械特性、導電性にとっては有利に作用するが、成形時の流動性の点では繊維は短い方が有利である。要求される導電性および成形性との兼ね合いから適宜選択すれば良い。通常押出機によるコンパウンドや射出成形等の成形工程によって、繊維は破損する。このため、成形前のペレット中のガラス繊維及び炭素繊維の繊維長及び繊維径としては、数平均繊維長

(L)と繊維径(d)の比(L/d)が10～10,000の範囲にあることが好ましく、更に好ましくは10～8,000である。また、成形品中におけるL/dとしては、5～5,000の範囲にあることが好ましく、更に好ましくは10～3,000である。

【0015】L/dが上記範囲であると、機械物性、導電性に優れる一方、成形時の流動性にも優れ、繊維同士が絡み合いにくく、分散が良好となる。

【0016】なお、上記の数平均繊維長はペレットまたは成形品を450℃×5時間、アルゴンガス雰囲気下において熱分解させて残った灰分から、1,000本の繊維について顕微鏡観察により測定した値から算出した値である。

【0017】本発明で使用される炭素繊維はチョップド繊維として押出機、ニーダー、パンバリーミキサーなどでカーボンナノチューブと熱可塑性樹脂と共に熔融混練しても良いし、ブルルージョン法のように炭素繊維の単繊維または繊維束を熔融した熱可塑性樹脂に含浸させながら引き取ったものをカーボンナノチューブと熱可塑性樹脂と共に熔融混練しても良い。あるいはスクリー及び/またはシリンダーの少なくとも1部を表面異形加工した開繊・繊維長制御機構部を有する押出機を使用し、この押出機に熱可塑性樹脂を供給して熔融させ、熔融樹脂中に連続状態の炭素繊維を供給して上記開繊・繊維長制御機構部で開繊・切断し、熔融樹脂中に均一分散させ、押出機から押出したものをカーボンナノチューブと熱可塑性樹脂と共に熔融混練しても良い。この方法では単繊維を集束したロービングが好ましく用いられる。集束本数は特に限定されず、単繊維のモノフィラメントを10～50000本集束したものが作業性の面で好ましく用いられる。この方法で用いられる押出機は、連続状態の繊維を開繊し、繊維長さを制御するようにスクリー及び/またはシリンダーを加工した単軸または多軸のスクリー式押出機であって、その内部に連続した繊維状強化材の開繊度や繊維長の制御機構部を有するものである。

【0018】本発明の樹脂組成物に配合される炭素繊維の重量には特に制限はないが、成形時の流動性、得られる成形品の比重及び強度、導電性の観点から樹脂組成物

全体の10~70重量%であることが好ましい。より好ましくは15~70重量%、更に好ましくは20~65重量%である。

【0019】本発明で使用するカーボンナノチューブは、炭素六角面が円筒状に閉じた単層構造あるいはこれらの円筒構造が入れ子状に配置された多層構造をした材料のことである。単層構造のみから構成されていても多層構造のみから構成されていても良く、単層構造と多層構造が混在していてもかまわない。また部分的にカーボンナノチューブの構造を有している炭素材料も使用で

【0020】カーボンナノチューブは、例えば炭素電極間にアーク放電を発生させ、放電用電極の陰極表面に成長させる方法、シリコンカーバイドにレーザービームを照射して加熱・昇華させる方法、遷移金属系触媒を用いて炭化水素を還元雰囲気下の気相で炭化する方法などによって製造することができる。製造方法の違いによって得られてくるカーボンナノチューブのサイズや形態は変わって来るが、いずれの形態のものも使用できる。

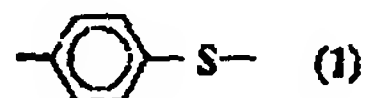
【0021】本発明の樹脂組成物に配合されるカーボンナノチューブの重量には特に制限は無いが、成形時の流動性、得られる成形品の比重及び強度、導電性の観点から樹脂組成物全体の0.1~15重量%であることが好ましい。より好ましくは0.5~10重量%、更に好ましくは1~8重量%である。

【0022】本発明で使用する熱可塑性樹脂の種類には特に制限は無いが、ポリオレフィン、ポリアミド、ポリアセタール、非液晶ポリエステル、ポリフェニレンオキッド、ポリカーボネート、ポリフェニレンスルフィド、液晶ポリエステル、ポリエーテルケトン、ポリエーテルエーテルケトン、ポリスルホン、ポリエーテルスルホン、ポリエーテルイミドおよびこれらのブレンド物からなる群から選ばれる一種が好ましく用いられる。これらの中でも流動性の面からポリフェニレンスルフィドおよび液晶ポリエステルが特に好ましい。

【0023】本発明で用いられるポリフェニレンスルフィド樹脂とは、一般式(1)で表される繰り返し単位を有する重合体であり、その含有量は耐熱性の面から70モル%以上であることが好ましく、より好ましくは80モル%以上、特に好ましくは90モル%以上である。繰り返し単位(1)の含有量が70モル%未満の場合には耐熱性および剛性が低下する傾向が見られる。一般式(1)以外の繰り返し単位としては、一般式(2)~(8)で表される構造単位が用いられる。

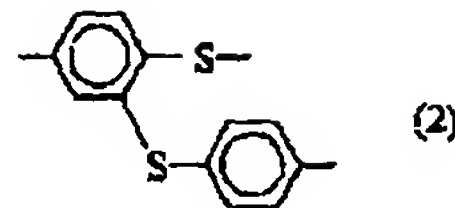
【0024】

【化1】



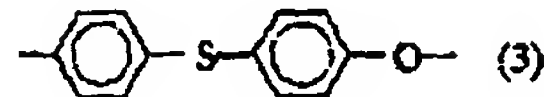
【0025】

【化2】



【0026】

【化3】



【0027】

【化4】



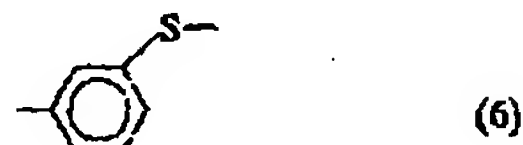
【0028】

【化5】



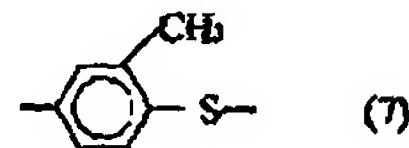
【0029】

【化6】



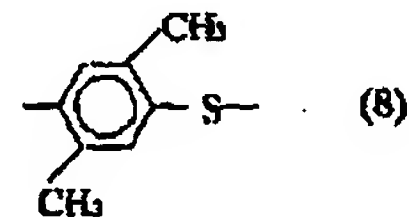
【0030】

【化7】



【0031】

【化8】



【0032】本発明で用いるポリフェニレンスルフィド樹脂としては、長さ31.75mm、径2.10mmのオリフィスを用い、温度316℃、荷重20gで測定した時のメルトフローレイト(以下MFRと略す)が50~200g/minであるものが好ましい。更に好ましくは50~150g/10min、とりわけ70~150g/10minが好ましい。

【0033】また、本発明で用いるポリフェニレンスルフィド樹脂の灰分は、450~500℃で炭化させた後、538℃で6時間灰化させた時の灰分残さ量で0.30重量%以下であることが好ましい。更に好ましくは0.25重量%以下、とりわけ0.22重量%以下が好ましい。

【0034】このようなポリフェニレンスルフィド樹脂は公知の方法、例えば特公昭45-3368号公報、特

公昭52-12240号公報、特開昭61-7332号公報に記載されている方法を用いて製造することができる。本発明においては、前記公報に記載されている方法で得られたポリフェニレンスルフィド樹脂を空气中加熱により架橋・高分子量化、窒素などのガス雰囲気下あるいは減圧下での熱処理、有機溶剤・熱水・酸性水溶液・アルカリ性水溶液などによる洗浄を施した上で使用しても良い。特に有機溶剤で洗浄すると低分子量成分が除去されるため、溶融成形時のガス発生、金型腐蝕は低減される。その場合の有機溶剤としては、N-メチルピロリドン、N, N'-ジメチルホルムアミド、N, N'-ジメチルアセトアミド、1, 3-ジメチルイミダゾリジノン、ヘキサメチルホスホンアミド、ビペラジノンなどの含窒素溶剤、ジメチルスルホキシド、ジメチルスルホン、スルホランなどのスルホン系溶剤、アセトン、メチルエチルケトン、ジエチルケトン、アセトフェノンなどのケトン系溶剤、ジメチルエーテル、ジエチルエーテル、ジブチルエーテル、1, 4-ジオキサン、テトラヒドロフランなどのエーテル系溶剤、クロロホルム、メチレンジクロリド、四塩化炭素、トリクロロエチレン、ジクロロエチレン、クロルベンゼンなどのハロゲン系溶剤、メタノール、エタノール、プロパノール、ブタノール、ペンタノール、エチレングリコール、プロピレングリコール、ポリエチレングリコール、ポリプロピレングリコールなどのアルコール系溶剤、フェノール、クレゾールなどのフェノール系溶剤、ベンゼン、トルエン、キシレンなどの芳香族炭化水素系溶剤、ヘンタン、ヘキサン、シクロヘキサン、ヘプタン、オクタンなどの飽和炭化水素系溶剤などが挙げられる。

【0035】また、酸性水溶液、アルカリ性水溶液については、ポリフェニレンスルフィド樹脂を分解する作用の無いものであれば特に制限は無く、酢酸、塩酸、硫酸、硝酸、有機カルボン酸、有機スルホン酸、各種水酸化アルカリ水溶液などが用いられる。

【0036】また、酸無水物、エポキシ基、イソシアネート基などの官能基を有する化合物で処理しても良い。

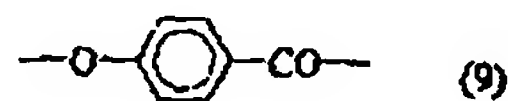
【0037】本発明で用いられる液晶ポリエステルは芳

香族オキシカルボニル単位、芳香族ジオキシ単位、脂肪族ジオキシ単位、芳香族ジカルボニル単位から選ばれる構造単位からなる。芳香族オキシカルボニル単位の実例としてはp-ヒドロキシ安息香酸、6-ヒドロキシ-2-ナフトエ酸から生成する構造単位、芳香族ジオキシ単位の実例としては4, 4'-ジヒドロキシジフェニル、ヒドロキノン、3, 3', 5, 5'-テトラメチル-4, 4'-ジヒドロキシビフェニル、1-ブチルヒドロキノン、フェニルヒドロキノン、2, 6-ジヒドロキシナフタレン、2, 7-ジヒドロキシナフタレン、2, 2-ビス(4-ヒドロキシフェニル)プロパン、4, 4'-ジヒドロキシジフェニルエーテルから生成する構造単位、脂肪族ジオキシ単位の実例としてはエチレングリコール、プロピレングリコールから生成する構造単位、芳香族ジカルボニル単位の実例としてはテレフタル酸、イソフタル酸、2, 6-ナフタレンジカルボン酸、4, 4'-ジフェニルジカルボン酸、1, 2-ビス(2-クロロフェノキシ)エタン-4, 4'-ジカルボン酸、4, 4'-ジフェニルエーテルジカルボン酸から生成する構造単位が挙げられる。

【0038】好ましい液晶ポリエステルは、構造単位(9)、(10)、(11)からなる液晶ポリエステルである。

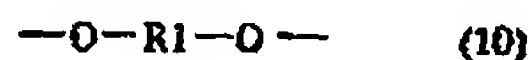
【0039】

【化9】



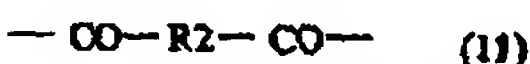
【0040】

【化10】



【0041】

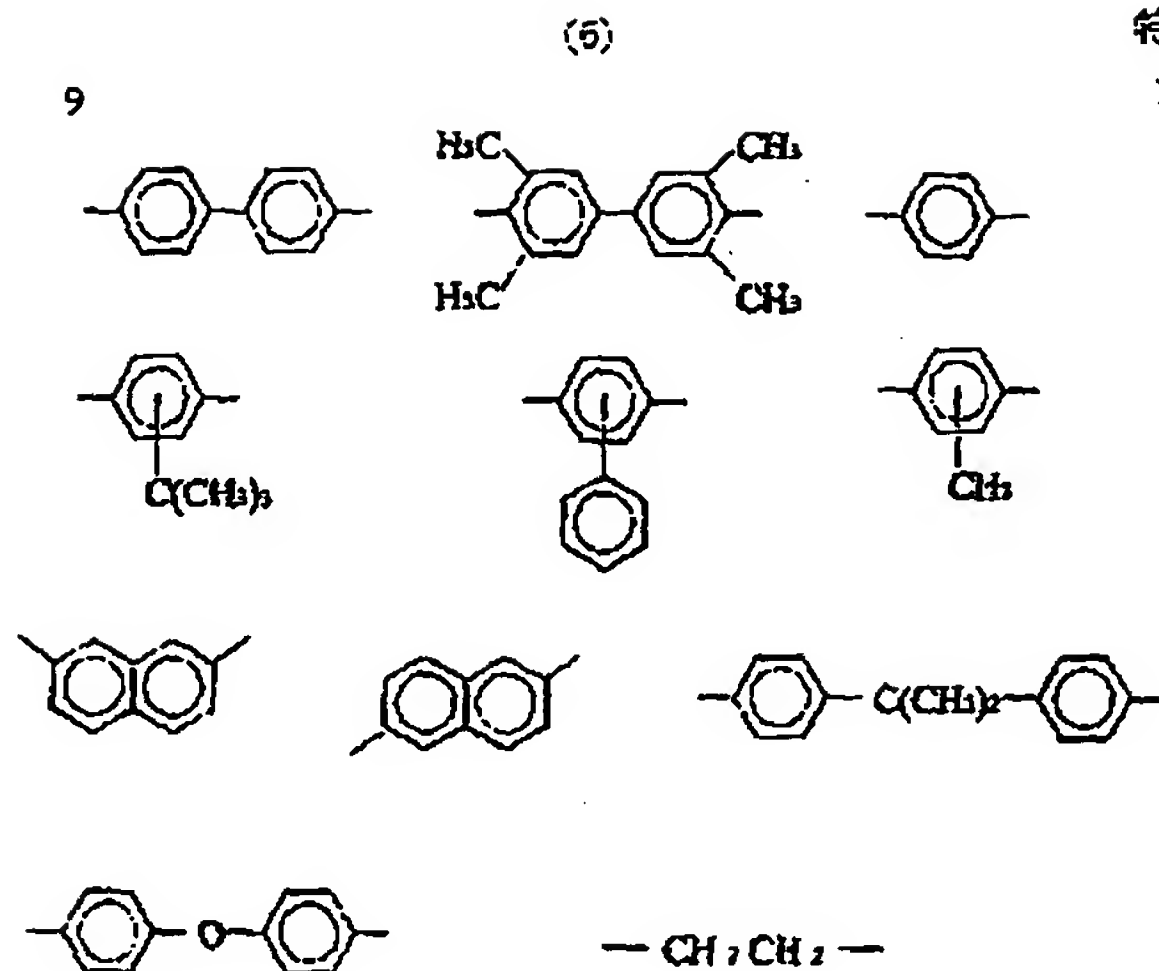
【化11】



【0042】ただし、式中R1は次の化学構造から選ばれた1種以上を表す。

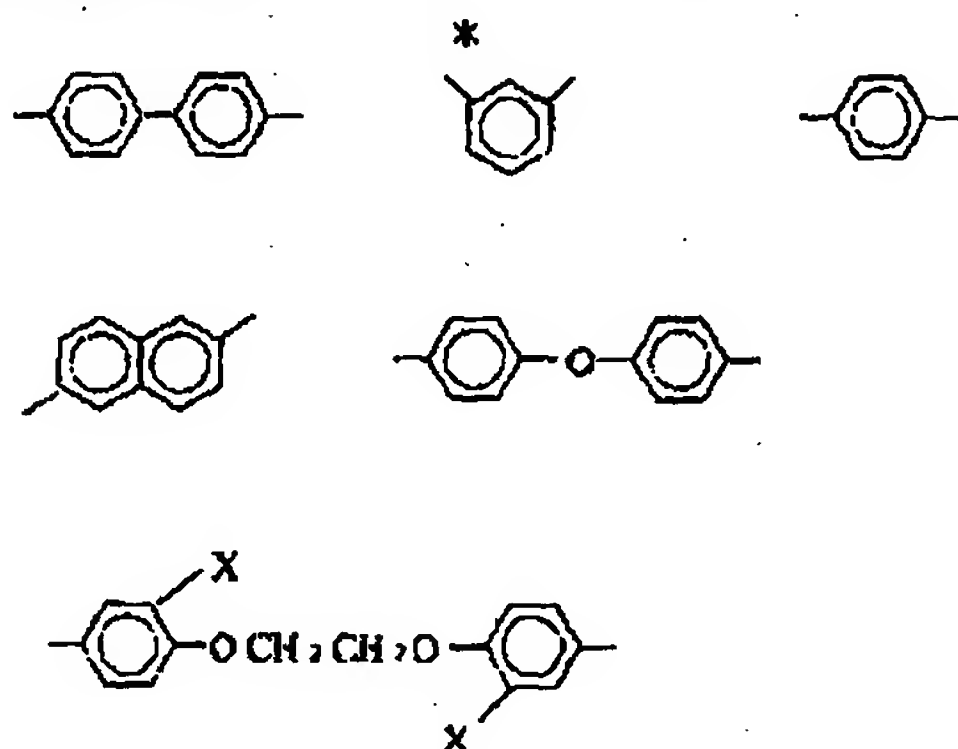
【0043】

【化12】



【0044】また式中R2は次の化学構造から選ばれた1種以上を表す。ただし、Xは水素原子またはハロゲン原子を表す。

*【0045】
【化13】



【0046】上記構造単位のうち、(10)についてはR1が4,4'-ビフェニレン、1,4-フェニレン、シメチレンであるものが好ましく、(11)についてはR2が1,4-フェニレン、2,6-ナフタレンであるものが好ましい。

【0047】本発明で好ましく用いられる液晶ポリエステルは上記の如く構造単位(9)、(10)、(11)からなる共重合体である。その共重合組成は任意に選ぶことができるが、次のような組成が好ましい。

【0048】上記構造単位(9)及び(10)の合計は構造単位(9)、(10)および(11)の合計に対して30~95mol%が好ましく、40~90mol%がより好ましい。また、構造単位(11)は構造単位(9)、(10)および(11)の合計に対して70~5mol%が好ましく、60~10mol%がより好ましい。また、構造単位(9)の構造単位(10)に対するmol比[(9)]/[(10)]は75/25~95/5であることが好ましく、より好ましくは78/22~93/7である。また、構造単位(11)は構造単位(10)と実質的に等モルであることが好ましい。こ

こで実質的に等モルとは、末端を除くポリマー主鎖を構成する構造単位が等モルであるが、末端を構成する構造単位としては必ずしも等モルとは限らないことを意味する。

【0049】本発明で好ましく用いられる上記液晶ポリエステルは上記構造単位(9)~(11)を構成する成分以外に、3,3'-ジフェニルジカルボン酸、2,2'-ジフェニルジカルボン酸などの芳香族ジカルボン酸、アジピン酸、アゼライン酸、セバシン酸、ドデカンジオン酸などの脂肪族ジカルボン酸、ヘキサヒドロテレフタル酸などの脂環式ジカルボン酸、クロロヒドロキノン、3,4'-ジヒドロキシジフェニル、4,4'-ジヒドロキシジフェニルスルホン、4,4'-ジヒドロキシジフェニルスルフィド、4,4'-ジヒドロキシベンゾフェノンなどのジフェノール、1,2-プロパンジオール、1,3-プロパンジオール、1,4-ブタンジオール、1,6-ヘキサンジオール、ネオペンチルグリコール、1,4-シクロヘキサジメタノールなどの脂肪族または脂環式ジオール、m-ヒドロキシ安息香酸、2,6-ヒドロキシナフトエ酸などの芳香族ヒドロキシ

カルボン酸などを液晶性を損なわない範囲で共重合せしめることができる。

【0050】液晶ポリエステル樹脂の対数粘度には特に制限はないが、ペンタフルオロフェノール中、0.1g/dL濃度、60℃で測定した値が0.5~15dL/gであることが好ましく、1~10dL/gであることが特に好ましい。

【0051】また、液晶ポリエステルの熔融粘度には特に制限はないが、流動性の面から融点+10℃の温度、ずり速度1,000sec⁻¹での値が0.5~200Pa・secであることが好ましく、0.5~100Pa・secであることが好ましい。なお、ここで言う融点(T_m)とは示差走査熱量測定において、室温から20℃/分の昇温条件で測定した時に観測される吸熱ピーク温度(T_{m1})を観測後、T_{m1}+20℃の温度で5分間保持した後、20℃/分の降温条件で室温まで冷却した後、再度20℃/分の昇温条件で昇温した時に観測される吸熱ピーク温度(T_{m2})のことを指す。

【0052】液晶ポリエステルの製造方法には特に制限がなく、公知のポリエステルの重縮合方法に従って製造することができる。例えば次のような方法で製造できる。

(1) p-ヒドロキシ安息香酸などのオキシカルボニル単位形成性成分を除く成分から得られたポリエステルとp-ヒドロキシ安息香酸とを乾燥窒素気流下で加熱溶解し、アシドリシス反応によって共重合ポリエステルフラグメントを生成させ、次いで減圧・増粘させる方法。

(2) p-アセトキシ安息香酸および4,4'-ジアセトキシジフェニル、ジアセトキシベンゼンなどの芳香族ジヒドロキシ化合物のジアシル化物と2,6-ナフタレンジカルボン酸、テレフタル酸、イソフタル酸などの芳香族ジカルボン酸から脱酢酸縮合反応させる方法。

(3) p-ヒドロキシ安息香酸および4,4'-ジヒドロキシジフェニル、ヒドロキノンなどの芳香族ジヒドロキシ化合物と2,6-ナフタレンジカルボン酸、テレフタル酸、イソフタル酸などの芳香族ジカルボン酸に無水酢酸を反応させてフェノール性水酸基をアシル化した後、脱酢酸重縮合反応する方法。

(4) p-ヒドロキシ安息香酸のフェニルエステル及び4,4'-ジヒドロキシジフェニル、ヒドロキノンなどのジフェノールと2,6-ナフタレンジカルボン酸、テレフタル酸、イソフタル酸などの芳香族ジカルボン酸のジフェニルエステルから脱フェノール重縮合反応により液晶ポリエステルの製造する方法。

(5) p-ヒドロキシ安息香酸および2,6-ナフタレンジカルボン酸、テレフタル酸、イソフタル酸などの芳香族ジカルボン酸に所定量のジフェニルカーボネートを反応させてそれぞれジフェニルエステルとした後、4,4'-ジヒドロキシジフェニル、ヒドロキノンなどのジフェノールを加え、脱フェノール重縮合反応により液晶

ポリエステルを製造する方法。

(6) ポリエチレンテレフタレートなどのポリエステルあるいはそのオリゴマーまたはビス(β-ヒドロキシエチル)テレフタレートなど芳香族ジカルボン酸のビス(β-ヒドロキシエチル)エステルの存在下で上記(2)または(3)の方法により液晶ポリエステルの製造する方法。

【0053】液晶ポリエステルの重縮合反応は無触媒でも進行するが、酢酸第1錫、テトラブチルチタネート、酢酸カリウム、酢酸ナトリウム、三酸化アンチモン、金属マグネシウムなどの金属化合物を使用することができる。

【0054】本発明で用いられる樹脂組成物には、本発明の効果を損なわない範囲で、ガラス繊維、チタン酸カリウムウイスカ、酸化亜鉛ウイスカ、酸化アルミニウムウイスカ、アラミド繊維、アルミナ繊維、炭化珪素繊維、セラミック繊維、アスベスト繊維、石こう繊維、金属繊維などの繊維状充填剤、ワラストナイト、ゼオライト、セリサイト、カオリン、マイカ、クレー、パイロフィライト、ペントナイト、アスベスト、タルク、アルミナシリケートなどの珪酸塩、アルミナ、酸化珪素、酸化マグネシウム、酸化シリコン、酸化チタン、酸化鉄などの金属化合物、炭酸カルシウム、炭酸マグネシウム、ドロマイトなどの炭酸塩、硫酸カルシウム、硫酸バリウムなどの硫酸塩、水酸化マグネシウム、水酸化カルシウム、水酸化アルミニウムなどの水酸化物、ガラスビーズ、ガラスフレーク、セラミックビーズ、窒化ホウ素、炭化珪素およびシリカなどの非繊維状充填剤が挙げられ、これらは中空であってもよく、さらにはこれら充填剤を2種類以上併用することも可能である。また、より優れた機械的強度を得る目的でこれら繊維状/非繊維状充填剤をイソシアネート系化合物、有機シラン系化合物、有機チタネート系化合物、有機ボラン系化合物、エポキシ化合物などのカップリング剤で予備処理して使用してもかまわない。

【0055】さらに、本発明の樹脂組成物には、タルク、カオリン、有機リン化合物、ポリエーテルエーテルケトンなどの結晶核剤、次亜リン酸塩などの着色防止剤、ヒンダードフェノール、ヒンダードアミンなどの酸化防止剤、熱安定剤、滑剤、紫外線防止剤、染料や顔料などの着色剤、帯電防止剤などの添加剤を添加することができる。

【0056】本発明の樹脂組成物の製造方法に特に制限はない。炭素繊維、カーボンナノチューブ、熱可塑性樹脂、その他成分を一括してドライブレンドした後、押出機、ニーダー、バンバリーミキサーなどで熔融混練しても良いし、予め熱可塑性樹脂と炭素繊維を熔融押出したペレットとカーボンナノチューブ、その他成分を熔融混練しても良い。あるいはブルトルージョン法で製造した炭素繊維/熱可塑性樹脂ペレットと予め熱可塑性樹脂と

カーボンナノチューブ、その他成分を溶融混練したペレットを更に溶融混練しても良い。

【0057】本発明の成形品の製造方法に特に制限は無く、射出成形、射出圧縮成形、プレス成形などを用いることができるが、生産効率の観点から射出成形が好ましい。炭素繊維、カーボンナノチューブ、熱可塑性樹脂、その他成分を一括してドライブレンドした後、そのまま射出成形しても良いし、一旦全成分を溶融押出してペレタイズしてから射出成形しても良い。あるいはブルトル

ーション法で製造した炭素繊維/熱可塑性樹脂ペレットと予め熱可塑性樹脂とカーボンナノチューブ、その他成分を溶融混練したペレットをドライブレンドしてそのまま射出成形しても良いし、両ペレットを一旦溶融押出してペレタイズしてから射出成形しても良い。

【0058】

【実施例】以下に実施例を示し、本発明を更に具体的に説明するが、本発明はこれら実施例の記載に限定されるものではない。

【0059】以下の実施例においてポリフェニレンスルフィド系材料の評価用試験片は、京芝機械1S80型射出成形機にて、シリンダー温度：320℃、金型温度：140℃、射出速度：75%、射出圧力：充填下限圧力+10kg/cm²(G)の設定条件で射出成形することにより作成した。

【0060】また、液晶ポリエステル系材料の評価用試験片は、京芝機械1S80型射出成形機にて、シリンダー温度：345℃、金型温度：120℃、射出速度：75%、射出圧力：充填下限圧力+10kg/cm²(G)の設定条件で射出成形することにより作成した。

【0061】以下の実施例において、機械特性、ガス透過性、体積固有抵抗の評価は次の方法により行った。

引張特性：ASTM D638に従って測定した。

曲げ特性：ASTM D790に従って測定した。

アイゾット衝撃強度：ASTM D256に従って測定した。

ガス透過性：水素ガスについては1mm厚みの試験片を使用し、JIS K7126に記載されているA法（差圧法）に従って23℃、飽和状態で測定した。また、水蒸気については25μm厚みの試験片を使用し、JIS

Z0208に従って40℃、90%RHで測定した。

体積固有抵抗：ASTM D257に従って測定した。

数平均繊維長(L)、繊維径(d)：ペレットまたは成形品を450℃×5時間、アルゴンガス雰囲気下において熱分解させて残った灰分から、1,000本の繊維について顕微鏡観察により測定して算出した。

【0062】【参考例1】ポリフェニレンスルフィド樹脂の製造

攪拌機付きオートクレーブに水酸化ナトリウム水溶液4.67kg（水酸化ナトリウム25モル）、50%水酸化ナトリウム2kg（水酸化ナトリウム25モル）な

らびにN-メチル-2-ピロリドン（以下NMPと略す。）8kgを仕込み、攪拌しながら徐々に昇温し、水3.8kgを含む蒸出水4.1Lを除去した。残留混合物に1,4-ジクロロベンゼン3.75kg（25.5モル）ならびにNMP2kgを加えて230℃で1時間加熱した。反応生成物を温水で5回洗浄後、90℃、pH4の酢酸水溶液25L中に投入し、1時間攪拌した。ポリフェニレンスルフィド樹脂を濾過し、濾液のpHが7になるまで90℃のイオン交換水で洗浄した後、80℃で24時間真空乾燥した。長さ31.75mm、径2.10mmのオリフィスを用い、温度316℃、荷重20gで測定した時のメルトフローレイト（MFR）は87g/min、450～500℃で炭化させた後、538℃で6時間灰化させた時の灰分残量は0.19重量%であった。

【0063】【参考例2】液晶ポリエステル樹脂の製造

p-ヒドロキシ安息香酸995重量部、4,4'-ジヒドロキシジフェニル126重量部、テレフタル酸112重量部、固有粘度が0.61dL/gのポリエチレンテレフタレート216重量部及び無水酢酸969重量部を攪拌器、蒸出管を備えた反応容器に仕込み、重合を行った。芳香族オキシカルボニル単位80mol%、芳香族ジオキシ単位7.5mol%、エチレンオキシ単位12.5mol%からなり、融点(Tm)314℃、0.5mmφ×10mmのオリフィスを用い、温度324℃、ずり速度1,000sec⁻¹で測定した時の熔融粘度が13Pa・secのペレットを得た。

【0064】【参考例3】径7mm、長さ48mmのグラファイト製スティックに、先端から中心軸に沿って径3mm、深さ29mmの穴を開け、この穴にロジウム；白金：グラファイト=5：5：2の混合粉末を詰めてカーボンナノチューブ製造用陽極を作成した。一方、99.998%純度のグラファイトからなる径14mm、長さ31mmの陰極を作成した。これらの電極を真空チャンバーの中に設置し、純度99.9%のヘリウムガスでチャンバー内部を置換し、直達アーク放電を行った。陽極と陰極の間隔を常に1～2mmに制御し、圧力600torr、電流70Aで放電を行った。陰極上に生成したカーボンナノチューブを取り出した。内径5nm、外径10nm、長さ1～10μmの単層及び複層のグラファイト層からなるカーボンナノチューブが得られた。

【0065】【実施例1～8】参考例1、参考例2で製造したポリフェニレンスルフィド樹脂及び液晶ポリエステル樹脂に直径7μmのPAN系炭素繊維（京レック「トレカ」T300）を数平均長さ6mmに切断したチョップド繊維及び参考例3で製造したカーボンナノチューブを表1に示す比率で配合し、30mmφの2軸押出機を用いてポリフェニレンスルフィド樹脂の場合は樹脂温度320℃、液晶ポリエステル樹脂の場合には樹脂温度3

40℃で、スクリー回転数150rpmで熔融混練し、樹脂組成物のペレットを製造した。このペレットを用いて射出成形により各種試験片を作製した。各種特性*

*の評価結果を表1に示す。
【0066】
【表1】

表1

実施例	実施例1	実施例2	実施例3	実施例4	実施例5	実施例6	実施例7	実施例8
炭素繊維 (wt%)	30	30	30	30	15	15	50	30
カーボンナノチューブ (wt%)	0.5	1	3	3	10	14	0.5	1
熱可塑性樹脂 [※] (種類)	PPS	PPS	PPS	PPS	PPS	PPS	PPS	LCP
(wt%)	69.5	69	67	65	75	71	49.5	69
引張強度 (MPa)	215	213	188	136	121	109	205	145
引張断面積 (%)	2.4	2.3	2.0	1.7	1.4	1.2	2.1	2.3
曲げ強度 (MPa)	259	251	216	182	142	128	291	192
曲げ弾性率 (GPa)	20.1	19.4	17.3	16.1	11.4	10.2	22.6	22.1
アイゾット衝撃値 (V/ft)	67	54	45	42	34	32	80	71
水蒸気透過性 (cm ³ /m ² ・atm・24h)	28	27	25	23	32	28	18	15
水素気透過性 (cm ³ /m ² ・atm・24h)	0.29	0.28	0.25	0.24	0.33	0.27	0.19	0.15
数平均繊維長/平均繊維径 (ペレット)	71	70	71	72	71	70	73	71
(成形品)	42	42	43	42	41	42	43	41
体積固有抵抗 (Ω・cm)	0.2	0.1	0.09	0.08	0.08	0.07	0.03	0.05

※ PPS：ポリフェニレンスルフィド樹脂、LCP：液晶ポリエステル樹脂

【0067】実施例1～8より本発明の組成物は機械的特性、ガスバリア性、導電性に優れ、かつ射出成形可能な材料であることがわかる。

【0068】実施例1～4より、炭素繊維の配合量を30wt%に固定し、カーボンナノチューブ量を増量すると機械的特性が若干低下し、導電性が若干上昇することがわかる。

【0069】実施例5～7より、炭素繊維とカーボンナノチューブの配合量を大きく変えても機械的特性、ガスバリア性、導電性に優れ、かつ射出成形可能な材料が得られるが、カーボンナノチューブを多量に配合しても導

電性の大幅な向上は見られず、機械的特性は低下する傾向が見られる。

【0070】実施例8より、マトリックス樹脂を変えても機械的特性、ガスバリア性、導電性に優れ、かつ射出成形可能な材料が得られることがわかる。

【0071】【比較例1～2】各成分の配合比率を表2のようにする以外は実施例1と同様に樹脂組成物を製造した。配合比率および特性評価結果を表2に示す。

【0072】

【表2】

表2

比較例	比較例1	比較例2	比較例3	比較例4
炭素繊維 (wt%)	30	—	黒鉛 76.8	ビッチ系炭素繊維 5
カーボンナノチューブ (wt%)	—	3	硬化剤 4	黒鉛 25 炭素性黒鉛 50
熱可塑性樹脂 *1 (種類)	PFS	PFS	エポキシ樹脂	LCP
(wt%)	70	97	19.2	20
引張強度 (MPa)	223	85	50	20
引張延伸率 (%)	2.5	13.2	1.3	0.9
曲げ強度 (MPa)	267	142	111	27
曲げ弾性率 (GPa)	20.2	3.7	4.1	2.1
アイゾット衝撃値 (V/ft)	65	45	44	4.1
水素ガス透過性 (cm ³ /m ² ・mm・24h)	28	46	53	52
水素気透過性 (cm ³ /m ² ・mm・24h)	0.28	0.48	0.88	0.86
数平均繊維長/平均繊維径 (μm)	70	—	—	—
(成形品)	42	—	—	—
体積固有抵抗 (Ω・cm)	149	1×10 ⁸	0.09	0.07

*1 PFS：ポリフェニレンスルフィド樹脂、LCP：液晶ポリエステル樹脂

【0073】比較例1～2より、カーボンナノチューブ又は炭素繊維の一方が欠けると機械的特性、ガスバリア性、導電性、射出成形性のすべてを満足する材料が得られないことがわかる。

【0074】【実施例9】実施例3で製造した組成物を用い、図1に示す形状の金型を使用して樹脂温度320℃、金型温度150℃、冷却時間20秒で連続射出成形を行った。連続1,000ショットを超えても不良の無い成形品が安定して得られた。図1は、燃料電池セパレーター用に水素ガス、酸素ガスまたは空気の流路を形成させた平板であり、サイズ66mm×84mm、厚み3mmである。当該平板には、ガス流路入り口A、ガス流路出口Bおよびガス流路Cが備えられている。また、ガス流路の幅は2.5mm、深さは2mmである。

【0075】【比較例3】炭素粉末として粒径110μm以上かつ160μm以下の黒鉛粉末をエポキシ樹脂（油化シェルエポキシ製“エピコート”807）に20：80重量部の比率で混合し、次いで硬化剤（油化シェルエポキシ製“エピキュア”1BM112）を4部添加して混合脱泡した。次いでこの材料を用い、温度150℃、圧力10MPaで45分間プレス成形して各種評価試験片を作成した。更に成形後に150℃で6時間キュアした後、物性を評価した。結果を表2に示す。

【0076】表2からわかるように、導電性には優れたものの、機械的特性に劣ることがわかる。

【0077】【比較例4】比較例3で調製した材料を用い、図1に示す形状の平板のプレス金型を使用して温度150℃、圧力10MPa、プレス時間45分で連続し

てプレス成形を行った。未充填、離型時破損などが発生し、10ショットを超える連続プレス成形が出来なかった。

【0078】【比較例5】参考例2で製造した液晶ポリエステル樹脂に膨張性黒鉛（住金ケミカル社製膨張性黒鉛TEG 80 LTE-U）、カーボンブラック（ケッチェンブラックインターナショナル社製ケッチェンブラックEC）、ビッチ系炭素繊維（三菱化学製ダイアリードK223QG）を表2に示す割合で配合し、溶融押出した。得られたペレットを用いて射出成形を試みたが成形できなかった。また、温度340℃、圧力10MPaで溶融プレス成形を行って各種評価試験片を作成し、物性を評価した。結果を表2に示す。表2からわかるように、導電性には優れたものの機械的特性に劣ることがわかる。

【0079】

【発明の効果】本発明を用いることにより、従来の方法では得られなかった成形性に優れた高度な導電性を有する樹脂組成物および成形品を得ることができる。また本発明の樹脂組成物より優れた燃料電池セパレーターを得ることができる。

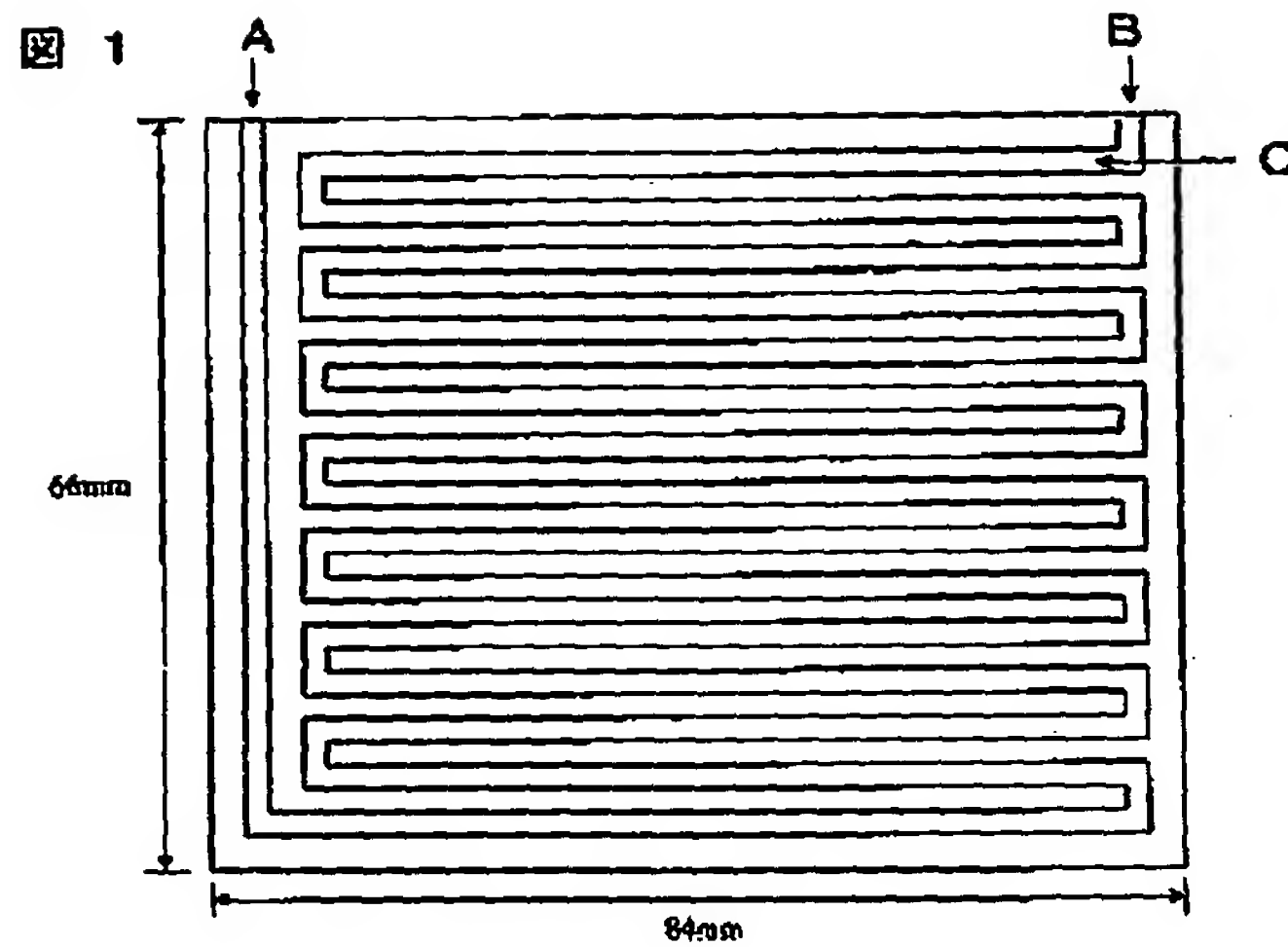
【図面の簡単な説明】

【図1】実施例9、比較例4で使用した平板の平面構造図である。

【符号の説明】

A：ガス流路入り口
B：ガス流路出口
C：ガス流路

【図1】



フロントページの続き

(51)Int.Cl.	識別記号	F I	フィート (参考)
H 0 1 M	8/02	H 0 1 M	8/02
// B 2 9 K	83:00	B 2 9 K	83:00
	105:12		105:12
B 2 9 L	31:00	B 2 9 L	31:00

F ターム (参考)

4F071 AA02 AA43 AA62 AB03 AD01
 AD06 AD07 AE15 AF08 AF37
 AH15 BA01 BB05 BC03
 4F206 AA24 AA34 AB18 AB25 AC07
 AE03 JA07 JF02
 4J002 AA011 BB001 CB001 CF001
 CG001 CH071 CH091 CL001
 CM041 CN011 CN031 DA016
 FA046 FA116 FD010 FD116
 G002
 5H026 AA02 CC03 EE05 EE18 HH01
 HH03 HH05

* NOTICES *

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CLAIMS

[Claim(s)]

[Claim 1] The thermoplastics constituent containing a carbon fiber and a carbon nanotube.

[Claim 2] The thermoplastics constituent according to claim 1 whose loadings of 10 - 70 % of the weight and a carbon nanotube the loadings of a carbon fiber are 0.1 - 15 % of the weight.

[Claim 3] The thermoplastics constituent according to claim 1 or 2 the number mean fiber length (L) of a carbon fiber and whose ratio (L/d) with the diameter of fiber (d) are 10-10,000.

[Claim 4] The thermoplastics constituent according to claim 1 to 3 which is more than a kind chosen from the group which thermoplastics becomes from a polyolefine, a polyamide, a polyacetal, non-liquid crystal polyester, a polyphenylene oxide, a polycarbonate, a polyphenylene sulfide, liquid crystal polyester, a polyether ketone, a polyether ether ketone, a polysulfone, polyether sulphone, polyether imide, and these blend objects.

[Claim 5] The thermoplastics constituent according to claim 1 to 3 whose thermoplastics is a polyphenylene sulfide or liquid crystal polyester.

[Claim 6] The thermoplastics constituent according to claim 1 to 3 whose thermoplastics is a polyphenylene sulfide.

[Claim 7] Mold goods which fabricate a thermoplastics constituent according to claim 1 to 6, and are obtained.

[Claim 8] Mold goods according to claim 7 the number mean fiber length (L) of the carbon fiber in mold goods and whose ratio (L/d) with the diameter of fiber (d) are 5-5,000.

[Claim 9] The separator for fuel cells which fabricates a thermoplastics constituent according to claim 1 to 6, and is obtained.

[Claim 10] The separator for fuel cells according to claim 9 the number mean fiber length (L) of the carbon fiber in mold goods and whose ratio (L/d) with the diameter of fiber (d) are 5-5,000.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the mold goods obtained from the thermoplastics constituent and it containing a carbon fiber and a carbon nanotube. Furthermore, it is related with the mold goods obtained from the thermoplastics constituent and it which were excellent in the conductivity and the moldability containing a carbon fiber and a carbon nanotube in detail.

[0002]

[Description of the Prior Art] Since a carbon-dioxide-gas eccrisis problem is coped with in recent years, the method of supplying energy by the method which does not depend the energy acquired from petroleum resources on petroleum resources is examined. A fuel cell is in one of them, and development is briskly performed aiming at spread to the power supply for fuel cell vehicles, or a power supply for home use.

[0003] Each cell of a fuel cell is formed from the separator which supplies a gas passageway and a cooling water way while it contacts one pair of electrodes and the electrode which sandwich the electrolyte which manages a reaction, and an electrolyte and performs current collection from an electrode.

[0004] Among these, since it has a current collection function from an electrode, advanced conductivity is required for a separator. Moreover, in order to form fuel gas passage and a circulating-water-flow way in both sides, gas barrier property, intensity, a corrosion resistance, etc. are required. Furthermore, it is necessary to form a gas passageway and a circulating-water-flow way, and the outstanding moldability is required.

[0005] After pressurizing the expanded-graphite particle and making a reserve sheet form as a conventional method, How to pressurize and produce with the metal mold of the last configuration where the gas passageway and the circulating-water-flow way were attached (JP,10-125337,A), A fluororesin or a graphite fluoride particle in a noble-metals matrix on a metal separator front face An eutectoid, How to form the distributed noble-metals compound plating coat (JP,2000-36309,A), How to blend a lot of graphites with thermosetting resin like phenol resin, and fabricate by press forming (JP,10-334927,A), The method (JP,2000-67882,A) of filling up precisely with conductive resin material the opening section of the metallic material which has the detailed opening section, the method (JP,2000-17179,A) of blending graphite system carbon material with a liquid crystal polymer so much, etc. are devised.

[0006]

[Problem(s) to be Solved by the Invention] however, a gas passageway fine by the method indicated by aforementioned JP,10-125337,A and a circulating-water-flow way -- metal mold -- since the top where it is difficult to form faithfully as a configuration runs short of water repellence and intensity, there is a trouble that the application of a high molecular compound, sinking in, etc. are needed Moreover, since the metal is used as main material by the method indicated by aforementioned JP,2000-36309,A, since a bird clapper and the coat of the material which uses noble metals as the base again are made to form, cost turns high up, a process becomes complicated, and a weight is not greatly realistic. furthermore, a gas passageway fine in press forming of the thermosetting resin which blended a lot of graphites by the method indicated by aforementioned JP,10-334927,A and a

circulating-water-flow way -- metal mold -- the trouble that a cycle time is long and inferior to productivity is in the top where it is difficult to form faithfully as a configuration By the method indicated by aforementioned JP,2000-67882,A, there was a trouble that an output declines according to generating of rust or elution of ion since the metallic material is used, or endurance fell. The trouble that the material strength of what can give advanced conductivity by the method indicated by aforementioned JP,2000-17179,A is inferior to a fluidity on a low, and inferior to productivity is *****.

[0007] Thus, by the conventional material and method, the material or the method for it being efficient and manufacturing cheaply the separator excellent in conductivity, gas barrier property, intensity, and the corrosion resistance were not found out.

[0008] Then, in this invention, are suitable for such a use and let it be a technical problem to offer material and mold goods excellent in gas barrier property, intensity, the corrosion resistance, and the moldability.

[0009]

[Means for Solving the Problem] Then, as a result of inquiring wholeheartedly that the above-mentioned technical problem should be solved, by using together and blending a carbon fiber and a carbon nanotube with thermoplastics, this invention persons found out that-izing could be carried out [high intensity] and advanced conductivity could be given, without spoiling the moldability of thermoplastics, and reached this invention.

[0010] Namely, the thermoplastics constituent with which this invention contains 1. carbon fiber and a carbon nanotube, 2. The thermoplastics constituent of one aforementioned publication whose loadings of 10 - 70 % of the weight and a carbon nanotube the loadings of a carbon fiber are 0.1 - 15 % of the weight, 3. The above 1 the number mean fiber length (L) of a carbon fiber and whose ratio (L/d) with the diameter of fiber (d) are 10-10,000, or the thermoplastics constituent of two publications, 4. Thermoplastics A polyolefine, a polyamide, a polyacetal, Non-liquid crystal polyester, a polyphenylene oxide, a polycarbonate, A polyphenylene sulfide, liquid crystal polyester, a polyether ketone, A polyether ether ketone, a polysulfone, polyether sulphone, A thermoplastics constituent given in either of the above 1-3 which is more than kinds chosen from the group which consists of polyether imide and these blend objects, 5. A thermoplastics constituent given in either of the above 1-3 whose thermoplastics is a polyphenylene sulfide or liquid crystal polyester, 6. A thermoplastics constituent given in either of the above 1-3 whose thermoplastics is a polyphenylene sulfide, 7. Mold goods which fabricate the thermoplastics constituent of a publication to either of the above 1-6, and are obtained, 8. Mold goods given in the above 7 the number mean fiber length (L) of the carbon fiber in mold goods and whose ratio (L/d) with the diameter of fiber (d) are 5-5,000, 9. The separator for fuel cells which fabricates the thermoplastics constituent of a publication to either of the above 1-6, and is obtained, 10. The number mean fiber length (L) of the carbon fiber in mold goods and a ratio (L/d) with the diameter of fiber (d) provide with the separator for fuel cells of a publication the above 9 which is 5-5,000.

[0011]

[Embodiments of the Invention] this invention is explained in detail below.

[0012] The carbon fiber used by this invention can use both the PAN system carbon fiber which uses the poly acrylic fiber as a raw material, and the pitch based carbon fiber which uses as a raw material the pitch which is a residue at the time of petroleum refining.

[0013] Although there is especially no limit about the path of the carbon fiber used by this invention, it is usually 1-30 micrometers, is 3-20 micrometers preferably, and is 5-15 micrometers still more preferably.

[0014] There is especially no limit in the length of the carbon fiber used by this invention. Although the longer one acts advantageously for a mechanical characteristic and conductivity, in respect of the fluidity at the time of fabrication, the shorter one of fiber is advantageous. What is necessary is just to choose from balance with the conductivity and the moldability which are demanded suitably. Usually, fiber is damaged by forming cycles by the extruder, such as a compound and injection molding. For this reason, as the fiber length and the diameter of fiber of the glass fiber in the pellet before fabrication, and a carbon fiber, it is 10-8,000 that the ratio (L/d) of number mean fiber length (L) and the diameter of fiber (d) is in the range of 10-10,000 desirable still more preferably.

Moreover, as L/d in mold goods, it is 10-3,000 that it is in the range of 5-5,000 desirable still more preferably.

[0015] While it excels that L/d is the above-mentioned range in machine physical properties and conductivity, it excels also in the fluidity at the time of fabrication, fiber cannot become entangled easily, and distribution becomes good.

[0016] In addition, the above-mentioned number mean fiber length is the value computed from the value measured by microscope observation about 1,000 fiber from the ash content which was made to pyrolyze a pellet or mold goods under argon gas atmosphere for 450 degree-Cx 5 hours, and remained.

[0017] The carbon fiber used by this invention may carry out melting kneading with a carbon nanotube and thermoplastics by the extruder, the kneader, the Banbury mixer, etc. as chopped fiber, and may carry out melting kneading of what was taken over while infiltrating the thermoplastics which fused the single fiber or fiber bundle of a carbon fiber like the protrusion rod method with a carbon nanotube and thermoplastics. Or the extruder which has opening and the fiber length controlling mechanism section which carried out surface variant processing of the at least 1 section of a screw and/or a cylinder is used, melting of the thermoplastics is supplied and carried out to this extruder, the carbon fiber of the successive state may be supplied into a melting resin, it may open and cut in the above-mentioned opening and fiber length controlling mechanism section, uniform distribution may be carried out into a melting resin, and melting kneading of what was extruded from the extruder may be carried out with a carbon nanotube and thermoplastics. By this method, roving which converged the single fiber is used preferably. Especially a number of filament in strand is not limited, but what converged the monofilament of a single fiber 10-50000 is preferably used in respect of workability. The extruder used by this method opens the fiber of the successive state, and as a fiber length is controlled, it is a screw-type extruder of the monopodium which processed the screw and/or the cylinder, or a multiple spindle, and it has the controlling mechanism section of the degree of opening of a fibrous reinforcement, or fiber length which followed the interior.

[0018] Although there is especially no limit in the weight of the carbon fiber blended with the resin constituent of this invention, it is desirable that they are a conductive viewpoint to the fluidity at the time of fabrication, the specific gravity of the mold goods obtained and intensity, and 10 - 70% of the weight of the whole resin constituent. It is 20 - 65 % of the weight still more preferably 15 to 70% of the weight more preferably.

[0019] The carbon nanotube used by this invention is the material to which the monolayer structures which the carbon hex-steel side closed in the shape of a cylinder, or such cylinder structures carried out multilayer structure arranged in the shape of a nest. It may consist of only multilayer structure, even if it consists of only monolayer structures, and it does not matter even if monolayer structure and multilayer structure are intermingled. Moreover, the carbon material which has the structure of a carbon nanotube partially can also be used. Moreover, it may be called by name called a graphite fibril nanotube besides a name called a carbon nanotube.

[0020] A carbon nanotube generates arc discharge for example, between carbon electrodes, and can be manufactured using heating, the method of making it sublimate, and a transition-metals system catalyst by the method of carbonizing a hydrocarbon by the gaseous phase under reducing atmosphere etc. by irradiating a laser beam at the method and silicon carbide which are grown up into the cathode surface of an electrode pattern. Anything of a form can be used although the size and the form of a carbon nanotube which are acquired by the difference in the manufacture method change.

[0021] Although there is especially no limit in the weight of the carbon nanotube blended with the resin constituent of this invention, it is desirable that they are a conductive viewpoint to the fluidity at the time of fabrication, the specific gravity of the mold goods obtained and intensity, and 0.1 - 15% of the weight of the whole resin constituent. It is 1 - 8 % of the weight still more preferably 0.5 to 10% of the weight more preferably.

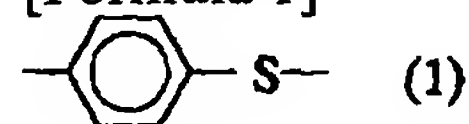
[0022] Although there is especially no limit in the kind of thermoplastics used by this invention, a kind chosen from the group which consists of a polyolefine, a polyamide, a polyacetal, non-liquid crystal polyester, a polyphenylene oxide, a polycarbonate, a polyphenylene sulfide, liquid crystal polyester, a polyether ketone, a polyether ether ketone, a polysulfone, polyether sulphone, polyether

imide, and these blend objects is used preferably. Especially a field to a fluid polyphenylene sulfide and fluid liquid crystal polyester are desirable also in these.

[0023] the polyphenylene-sulfide resin used by this invention is a polymer which has the repeat unit expressed with a general formula (1), and the content is more than 70 mol % from a heat-resistant field -- desirable -- more -- desirable -- more than 80 mol % -- it is more than 90 mol % especially preferably When the content of a repeat unit (1) is less than [70 mol %], the inclination for thermal resistance and rigidity to fall is seen. As repeat units other than a general formula (1), the structural unit expressed with general formula (2) - (8) is used.

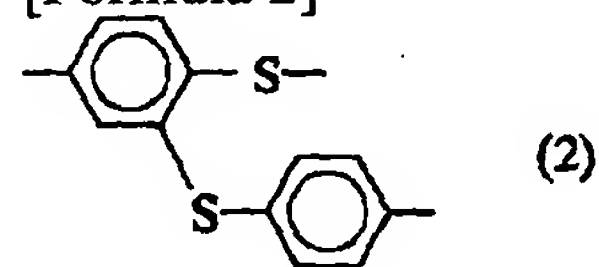
[0024]

[Formula 1]



[0025]

[Formula 2]



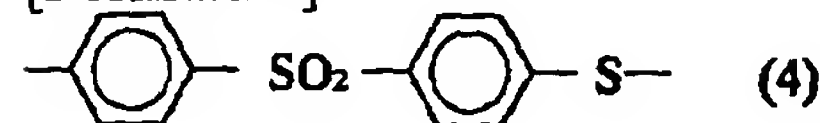
[0026]

[Formula 3]



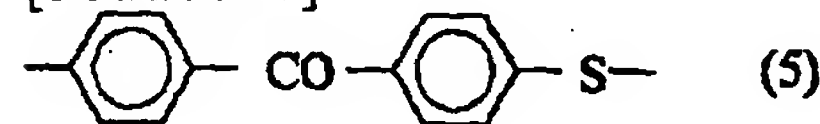
[0027]

[Formula 4]



[0028]

[Formula 5]



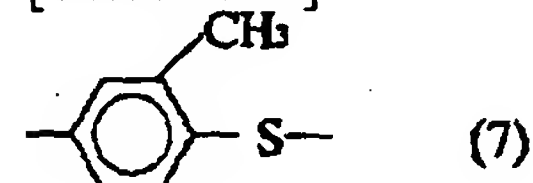
[0029]

[Formula 6]



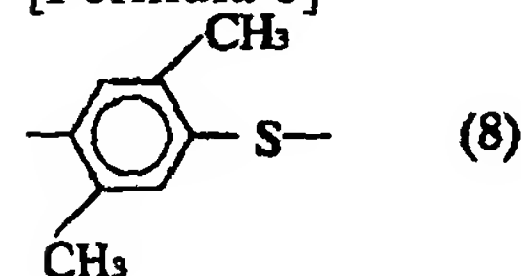
[0030]

[Formula 7]



[0031]

[Formula 8]



[0032] That whose melt flow rate (it omits Following MFR) when measuring by the temperature of 316 degrees C and 20g of loads, using a length of 31.75mm and the orifice of 2.10mm of diameters as a polyphenylene-sulfide resin used by this invention is 50 - 200 g/min is desirable. furthermore --

desirable -- 50-150g/-- 70-150g/10min is especially desirable 10 min

[0033] Moreover, after making it carbonize at 450-500 degrees C, as for the ash content of the polyphenylene-sulfide resin used by this invention, it is desirable that it is 0.30 or less % of the weight in the amount of ash content residues at the time of making it ash at 538 degrees C for 6 hours. Furthermore, 0.22 or less % of the weight is especially preferably desirable 0.25 or less % of the weight.

[0034] Such a polyphenylene-sulfide resin can be manufactured using the method indicated by a well-known method, for example, JP,45-3368,B, JP,52-12240,B, and JP,61-7332,A. In this invention, after performing washing according the polyphenylene-sulfide resin obtained by the method indicated by the aforementioned official report to heat treatment under gas atmosphere, such as crosslinked-polymer quantification and nitrogen, or reduced pressure, the organic solvent, hot water, acid solution, alkaline solution, etc. by heating among air, you may use it. since a low molecular weight constituent will be removed if it washes especially by the organic solvent -- the generation of gas at the time of melting fabrication, and metal mold -- corrosion is reduced As an organic solvent in that case, an N-methyl pyrrolidone, N, and N'-dimethylformamide, An N and N'-dimethylacetamide, 1, 3-dimethyl imidazolidinone, Nitrogen-containing solvents, such as a hexa methyl HOSUHON amide and piperazinone, dimethyl sulfoxide, Sulfone system solvents, such as a dimethyl sulfone and a sulfolane, an acetone, a methyl ethyl ketone, Ketone system solvents, such as a diethyl ketone and an acetophenone, a wood ether, Ethers solvents, such as diethylether, the dipropyl ether, 1, 4-dioxane, and a tetrahydrofuran, Chloroform, methylene dichloride, a carbon tetrachloride, a trichloroethylene, Halogen system solvents, such as a dichloroethylene and chlorobenzene, a methanol, Ethanol, propanol, a butanol, a pentanol, ethylene glycol, Alcohols solvents, such as a propylene glycol, a polyethylene glycol, and a polypropylene glycol, Saturated-hydrocarbon system solvents, such as aromatic hydrocarbons solvents, such as phenol system solvents, such as a phenol and cresol, benzene, toluene, and a xylene, a pentane, a hexane, a cyclohexane, a heptane, and an octane, etc. are mentioned.

[0035] Moreover, about acid solution and alkaline solution, if there is no operation which disassembles a polyphenylene-sulfide resin, there will be especially no limit and an acetic acid, a hydrochloric acid, a sulfuric acid, phosphoric acid, an organic carboxylic acid, an organic sulfonic acid, various hydroxylation alkali solution, etc. will be used.

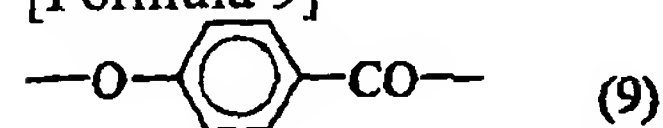
[0036] Moreover, you may process with the compound which has functional groups, such as an acid anhydride, an epoxy group, and an isocyanate machine.

[0037] The liquid crystal polyester used by this invention consists of a structural unit chosen from an aromatic oxy-carbonyl unit, an aromatic dioxy unit, an aliphatic dioxy unit, and an aromatic dicarbonyl unit. As an example of an aromatic oxy-carbonyl unit, para hydroxybenzoic acid, As an example of the structural unit generated from a 6-hydroxy-2-naphthoic acid, and an aromatic dioxy unit, 4 and 4'-dihydroxydiphenyl, A hydroquinone, 3, 3', 5, 5'-tetramethyl - 4 4'-dihydroxy biphenyl, t-butyl hydroquinone, a phenyl hydroquinone, 2, 6-dihydroxy naphthalene, 2, 7-dihydroxy naphthalene, 2, and 2-screw (4-hydroxyphenyl) propane, The structural unit generated from a 4 and 4'-dihydroxy diphenyl ether, The structural unit generated from ethylene glycol and a propylene glycol as an example of an aliphatic dioxy unit, As an example of an aromatic dicarbonyl unit, a terephthalic acid, an isophthalic acid, The structural unit generated from a 2, 6-naphthalene dicarboxylic-acid, 4, and 4'-diphenyl dicarboxylic acid, 1, 2-screw (2-chloro phenoxy) ethane -4, and a - dicarboxylic-acid, and 4 '4, 4'-diphenyl-ether dicarboxylic acid is mentioned.

[0038] Desirable liquid crystal polyester is liquid crystal polyester which consists of a structural unit (9), (10), and (11).

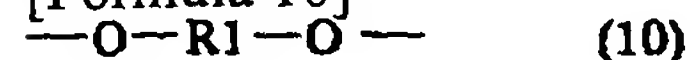
[0039]

[Formula 9]



[0040]

[Formula 10]



[0041]

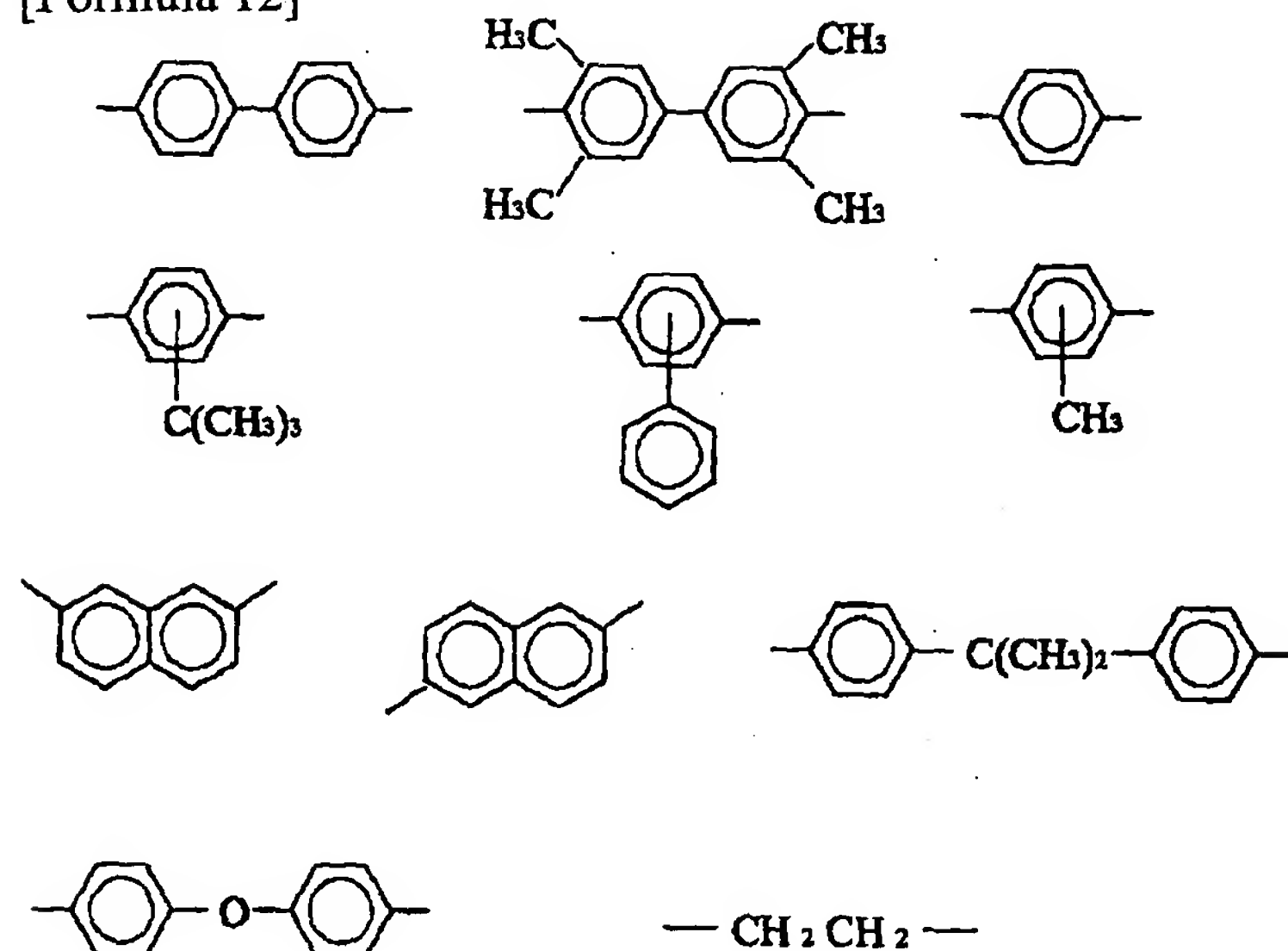
[Formula 11]



[0042] However, the inside R1 of a formula expresses one or more sorts chosen from the following chemical structure.

[0043]

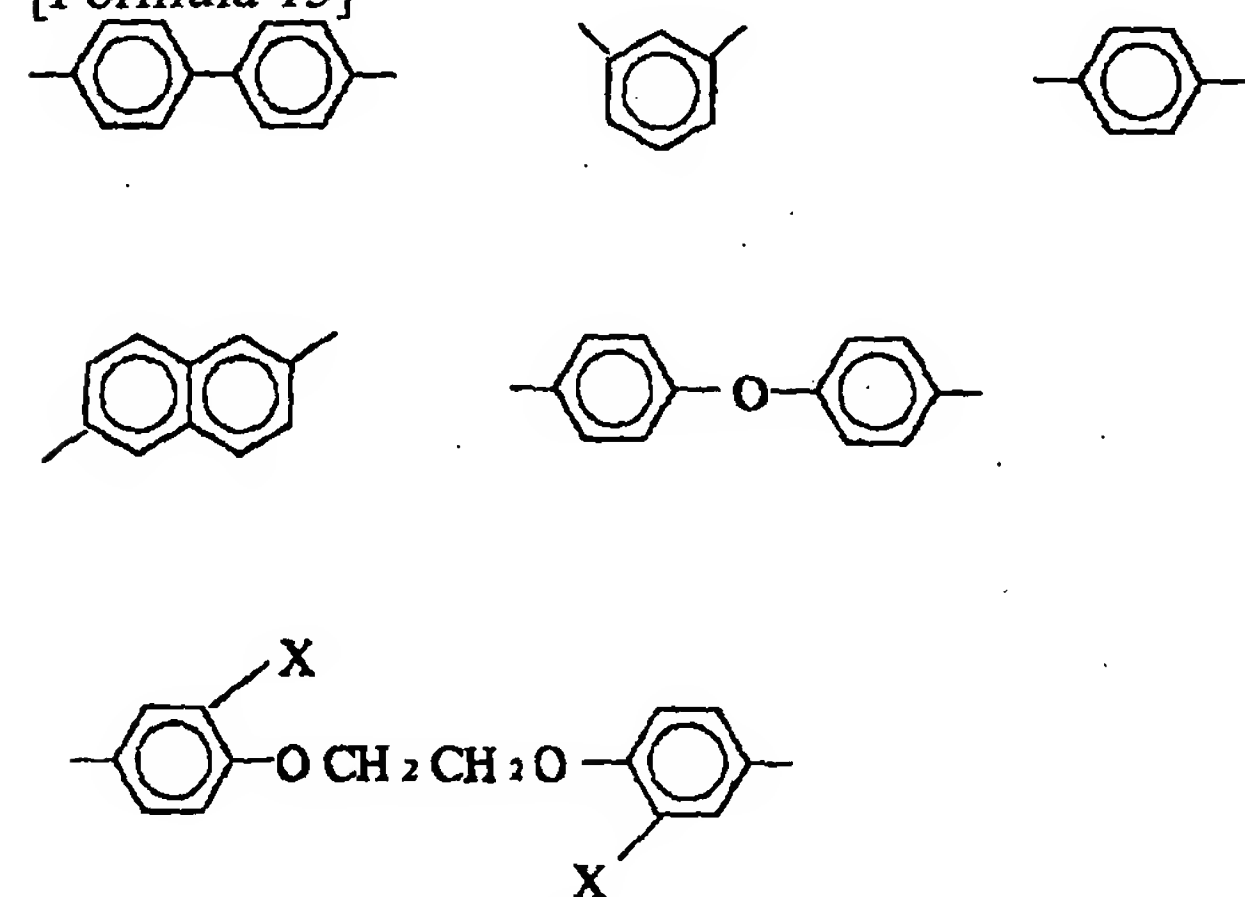
[Formula 12]



[0044] Moreover, the inside R2 of a formula expresses one or more sorts chosen from the following chemical structure. However, X expresses a hydrogen atom or a halogen atom.

[0045]

[Formula 13]



[0046] About (10), that whose R1 is 4 and 4'-biphenylene, 1, 4-phenylene, and dimethylene is desirable among the above-mentioned structural units, and that whose R2 is 1, 4-phenylene, 2, and 6-naphthalene is desirable about (11).

[0047] The liquid crystal polyester preferably used by this invention is a copolymer which consists of a structural unit (9), (10), and (11) like the above. The following composition is desirable although the copolymerization composition can be chosen arbitrarily.

[0048] The above-mentioned structural unit (9) and 30 - 95-mol% of the sum total of (10) are desirable to the sum total of a structural unit (9), (10), and (11), and are more desirable. [40 - 90 mol% of] Moreover, 70 - five-mol% of a structural unit (11) is desirable to the sum total of a structural unit (9), (10), and (11), and is more desirable. [60 - ten-mol% of] moreover -- a structural unit -- (-- nine --) -- a structural unit -- (-- ten --) -- receiving -- mol -- a ratio -- [-- (-- nine --) --] -- /-- [-- (-- ten --) --] -- 75 -- /-- 25 - 95 -- /-- five -- it is -- things -- desirable -- more -- desirable -- 78 / 22

- 93/7 -- it is . Moreover, as for a structural unit (11), it is desirable that they are mols [target / real / a structural unit (10),]. Although mols [target / real] are mols / structural unit / which constitutes the polymer principal chain except an end], they mean that it is not not necessarily a ** mol as a structural unit which constitutes an end here.

[0049] In addition to the component which constitutes above-mentioned structural-unit (9) - (11), the above-mentioned liquid crystal polyester preferably used by this invention Aromatic dicarboxylic acids, such as a - diphenyl dicarboxylic-acid, and 3 and 3 '2, 2'-diphenyl dicarboxylic acid, Aliphatic dicarboxylic acids, such as an adipic acid, an azelaic acid, a sebacic acid, and a dodecane dione acid, Alicyclic dicarboxylic acids, such as a hexahydro terephthalic acid, a chlorohydroquinone, 3, - dihydroxydiphenyl, and 4 '4, 4'-dihydroxy diphenylsulfone, Diphenols, such as a - dihydroxydiphenyl sulfide, and 4 and 4 '4, 4'-dihydroxy benzophenone, 1, 2-propanediol, 1, 3-propanediol, 1, 4-butanediol, Aliphatic series or alicyclic diols, such as 1, 6-hexandiol, neopentyl glycol, 1, and 4-cyclohexane dimethanol, Aromatic hydroxycarboxylic acid, such as an m-hydroxybenzoic-acid, 2, and 6-hydroxy naphthoic acid, etc. can be made to copolymerize in the range which does not spoil mesomorphism.

[0050] the logarithm of liquid crystal polyester resin -- although there is especially no limit in viscosity, among a pentafluoro phenol, it is desirable that 0.1 g/dL concentration and the value measured at 60 degrees C are 0.5 - 15 dL/g, and it is desirable that it is especially 1 - 10 dL/g

[0051] Moreover, although there is especially no limit in the melt viscosity of liquid crystal polyester, it is desirable that the temperature of +10 degrees C of a fluid field to melting points and the value of shear rate 1,000sec-1 are 0.5 - 200 Pa-sec, and it is desirable that it is 0.5 - 100 Pa-sec. In addition, after observing the endothermic peak temperature (Tm1) observed when it measures on 20-degree-C temperature up conditions for /from a room temperature in differential scanning calorimetry with the melting point (Tm) said here, The thing of the endothermic peak temperature (Tm2) observed when a temperature up is again carried out on 20-degree-C temperature up conditions for /after holding for 5 minutes at the temperature of 1+20 degree C of Tm(s), and cooling to a room temperature on 20-degree-C temperature fall conditions for /is pointed out.

[0052] There is no limit especially in the manufacture method of liquid crystal polyester, and it can manufacture according to the polycondensation method of well-known polyester. For example, it can manufacture by the following methods.

- (1) How to carry out heating fusion of the polyester and para hydroxybenzoic acid which were obtained from the component except oxy-carbonyl unit formation nature components, such as para hydroxybenzoic acid, under a dryness nitrogen air current, make generate copolymerized polyester fragmentation by the acidolysis reaction, and make it decompress and thicken subsequently.
- (2) How to carry out a ***** condensation polymerization reaction from aromatic dicarboxylic acids, such as a diacyl ghost of aromatic dihydroxy compounds, such as p-acetoxy benzoic-acid and 4, and 4'-diacetoxy diphenyl and diacetoxy benzene, 2 and 6-naphthalene dicarboxylic acid, a terephthalic acid, and an isophthalic acid.
- (3) How to carry out a ***** polycondensation reaction, after making an acetic anhydride react to aromatic dicarboxylic acids, such as aromatic dihydroxy compounds, such as para-hydroxybenzoic-acid and 4, and 4'-dihydroxydiphenyl and a hydroquinone, 2 and 6-naphthalene dicarboxylic acid, a terephthalic acid, and an isophthalic acid, and acylating a phenolic hydroxyl group.
- (4) How to manufacture liquid crystal polyester by the ** phenol polycondensation reaction from the diphenyl ester of aromatic dicarboxylic acids, such as diphenols, such as phenyl ester [of para hydroxybenzoic acid] and 4, and 4'-dihydroxydiphenyl, and a hydroquinone, 2 and 6-naphthalene dicarboxylic acid, a terephthalic acid, and an isophthalic acid.
- (5) How to add diphenols, such as 4 and 4'-dihydroxydiphenyl and a hydroquinone, and to manufacture liquid crystal polyester by the ** phenol polycondensation reaction after making the diphenyl carbonate of the specified quantity react to aromatic dicarboxylic acids, such as para hydroxybenzoic acid and 2, 6-naphthalene dicarboxylic acid, a terephthalic acid, and an isophthalic acid, and considering as diphenyl ester, respectively.
- (6) How to manufacture liquid crystal polyester by the above (2) or the method of (3) under existence of the screw (beta-hydroxyethyl) ester of an aromatic dicarboxylic acid, such as polyester, its oligomer, or screw (beta-hydroxyethyl) terephthalate, such as a polyethylene terephthalate.

[0053] Although the polycondensation reaction of liquid crystal polyester advances also with a non-catalyst, metallic compounds, such as the 1st tin of an acetic acid, tetrabutyl titanate, potassium acetate, sodium acetate, an antimony trioxide, and metal magnesium, can be used.

[0054] In the resin constituent used by this invention, it is a book. A glass fiber, a potassium-titanate whisker, a zinc-oxide whisker, a boric-acid aluminum whisker, An aramid fiber, an alumina fiber, a silicon carbide fiber, a ceramic fiber, asbestos fiber, Fibrous bulking agents, such as stone KOU fiber and a metal fiber, a WARASUTE night, a zeolite, A sericite, a kaolin, a mica, clay, a pyrophyllite, a bentonite, Silicate, such as asbestos, talc, and an alumina silicate, an alumina, Metallic compounds, such as oxidization silicon, a magnesium oxide, a zirconium oxide, titanium oxide, and an iron oxide, Carbonates, such as a calcium carbonate, a magnesium carbonate, and a dolomite, a calcium sulfate, Sulfates, such as a barium sulfate, a magnesium hydroxide, a calcium hydroxide, Un-fibrous bulking agents, such as hydroxides, such as an aluminum hydroxide, a glass bead, glass flakes, a ceramic bead, boron nitride, a silicon carbide, and a silica, may be mentioned, these may be hollow, and it is also possible to use together two or more kinds of these bulking agents further. Moreover, you may use fibrous / [these] un-fibrous filler by coupling agents, such as an isocyanate system compound, an organic silane system compound, an organic titanate compound, an organic borane system compound, and an epoxy compound, carrying out preliminary processing in order to obtain the more excellent mechanical strength.

[0055] Furthermore, to the resin constituent of this invention, additives, such as coloring agents, such as antioxidants, such as coloring inhibitors, such as crystalline-nucleus agents, such as talc, a kaolin, an organic phosphorous compound, and a polyether ether ketone, and hypophosphite, a hindered phenol, and hindered amine, a thermostabilizer, lubricant, an ultraviolet-rays inhibitor, a color, and a pigment, and an antistatic agent, can be added.

[0056] There is especially no limit in the manufacture method of the resin constituent of this invention. After carrying out the dryblend of a carbon fiber, a carbon nanotube, thermoplastics, and the other components collectively, you may carry out melting kneading of the pellet which could carry out melting kneading by the extruder, the kneader, the Banbury mixer, etc., and carried out melting extrusion of thermoplastics and the carbon fiber beforehand, a carbon nanotube, and the other components. Or you may carry out melting kneading of the carbon fiber / thermoplastics pellet manufactured by the protrusion rod method, and the pellet which carried out melting kneading of thermoplastics, a carbon nanotube, and the other components beforehand further.

[0057] Although there is especially no limit in the manufacture method of the mold goods of this invention and injection molding, injection compression molding, press forming, etc. can be used, the viewpoint of productive efficiency to injection molding is desirable. You may carry out injection molding, once it may carry out injection molding as it is, and carries out melting extrusion of all the components and pelletizes them, after carrying out the dryblend of a carbon fiber, a carbon nanotube, thermoplastics, and the other components collectively. Or the dryblend of the carbon fiber / thermoplastics pellet manufactured by the protrusion rod method, and the pellet which carried out melting kneading of thermoplastics, a carbon nanotube, and the other components beforehand may be carried out, and it may carry out injection molding as it is, and after once carrying out melting extrusion of both the pellets and pelletizing them, you may carry out injection molding.

[0058]

[Example] Although an example is shown below and this invention is explained to it still more concretely, this invention is not limited to the publication of these examples.

[0059] In the following examples, the test piece for evaluation of polyphenylene-sulfide system material was created by carrying out injection molding by the setups of injection-pressure:restoration minimum pressure +10 kg/cm² (G) cylinder-temperature:320 degree C, die-temperature:140 degree C, and injection-speed:75% with the Toshiba Machine information separator 80 type injection molding machine.

[0060] Moreover, the test piece for evaluation of liquid crystal polyester system material was created by carrying out injection molding by the setups of injection-pressure:restoration minimum pressure +10 kg/cm² (G) cylinder-temperature:345 degree C, die-temperature:120 degree C, and injection-speed:75% with the Toshiba Machine information separator 80 type injection molding machine.

[0061] In the following examples, evaluation of a mechanical characteristic, gas permeability, and

volume resistivity was performed by the following method.

Tractive characteristics: ASTM It measured according to D638.

Bending property: ASTM It measured according to D790.

Izod impactive strength: ASTM It measured according to D256.

Gas permeability: Use the test piece of 1mm thickness about hydrogen gas, and it is JIS. According to the A method (differential pressure method) indicated by K7126, it measured by 23 degrees C and the absolute dry condition. Moreover, the test piece of 25-micrometer thickness is used about a steam, and it is JIS. According to Z0208, it measured by 40 degrees C and 90%RH.

Volume resistivity: ASTM It measured according to D257.

Number mean fiber length (L), the diameter of fiber (d): It computed by having measured by microscope observation about 1,000 fiber from the ash content which was made to pyrolyze a pellet or mold goods under argon gas atmosphere for 450 degree-Cx 5 hours, and remained. [0062] [Example 1 of reference] The temperature up was carried out gradually, having taught 8kg (it omitting Following NMP.) of N-methyl-2-pyrrolidones at 4.67kg (25 mols of specific hydrosulfides) of specific-hydrosulfide solution, and 2kg (25 mols of sodium hydroxides) row of 50% sodium hydroxides, and stirring to the autoclave with a manufacture agitator of a polyphenylene-sulfide resin, and distillate water 4.1L containing 3.8kg of water was removed. 1 was added to remains mixture, NMP2kg was added to the 4-dichlorobenzene 3.75kg (25.5 mols) row, and it heated at 230 degrees C for 1 hour. With warm water, the resultant was supplied after 5 times washing and in 90 degrees C and acetic-acid solution 25L of pH 4, and was stirred for 1 hour. The polyphenylene-sulfide resin was filtered, and after washing by 90-degree C ion exchange water until pH of filtrate was set to 7, the vacuum drying was carried out at 80 degrees C for 24 hours. After carbonizing the melt flow rate (MFR) when measuring by the temperature of 316 degrees C, and 20g of loads using a length of 31.75mm, and the orifice of 2.10mm of diameters at 87 g/min and 450-500 degrees C, the amount of ash content residues at the time of making it ash at 538 degrees C for 6 hours was 0.19 % of the weight.

[0063] [Example 2 of reference] The manufacture para-hydroxybenzoic-acid 995 weight section [of liquid crystal polyester resin], 4, and 4'-dihydroxydiphenyl 126 weight section, the terephthalic-acid 112 weight section, and intrinsic viscosity taught the polyethylene-terephthalate 216 weight section of 0.61 dL/g, and the acetic-anhydride 969 weight section to the reaction container equipped with stirring wings and the distillate pipe, and the polycondensation was performed. It consisted of 12.5 mol % of 7.5 mol % and ethyleneoxy units of 80 mol % and aromatic dioxy units of aromatic oxy-carbonyl units, and the melt viscosity when measuring by the temperature of 324 degrees C and shear rate 1,000sec⁻¹ obtained the pellet of 13 Pa-sec using melting point (Tm)314 degree C and the 0.5mm phix10mm orifice.

[0064] On 7mm of diameters of [the example 3 of reference], and the stick made from graphite with a length of 48mm, along with the medial axis, 3mm of diameters and the hole with a depth of 29mm were made from the nose of cam, the mixed-powder end of rhodium:platinum:graphite=5:5:2 was put in this hole, and the anode plate for carbon nanotube manufacture was created. On the other hand, 14mm of diameters and the cathode with a length of 31mm which consist of graphite of purity 99.998% were created. These electrodes were installed into the vacuum chamber, the interior of a chamber was replaced by gaseous helium of 99.9% of purity, and DC arc electric discharge was performed. The interval of an anode plate and cathode was always controlled to 1-2mm, and it discharged by pressure 600torr and current 70A. The carbon nanotube generated on cathode was taken out. The carbon nanotube which consists of a graphite layer of a monolayer with the bore of 5nm, an outer diameter [of 10nm], and a length of 1-10 micrometers and a double layer was obtained.

[0065] The carbon nanotube which manufactured the PAN system carbon fiber (Toray Industries make "TOREKA" T300) with a diameter of 7 micrometers in the chopped fiber and the example 3 of reference which were cut in number-average length of 6mm to the polyphenylene-sulfide resin and liquid crystal polyester resin which were manufactured in the example 1 of [examples 1-8] reference, and the example 2 of reference It blended by the ratio shown in Table 1, and using the biaxial extruder of 30mmphi, in the case of the polyphenylene-sulfide resin, in the case of the resin temperature of 320 degrees C, and liquid crystal polyester resin, it is 340 degrees C in resin

temperature, melting kneading was carried out by screw-speed 150rpm, and the pellet of a resin constituent was manufactured. Various test pieces were produced with injection molding using this pellet. The evaluation result of various properties is shown in Table 1.

[0066]

[Table 1]

表 1

実施例	実施例 1	実施例 2	実施例 3	実施例 4	実施例 5	実施例 6	実施例 7	実施例 8
炭素繊維 (wt%)	30	30	30	30	15	15	50	30
カーボンナノチューブ (wt%)	0.5	1	3	5	10	14	0.5	1
熱可塑性樹脂 *1 (種類)	PPS	PPS	PPS	PPS	PPS	PPS	PPS	LCP
(wt%)	69.5	69	67	65	75	71	49.5	69
引張強度 (MPa)	215	213	188	156	121	109	205	145
引張破断伸び (%)	2.4	2.3	2.0	1.7	1.4	1.2	2.1	2.3
曲げ強度 (MPa)	259	251	216	182	142	128	291	192
曲げ弾性率 (GPa)	20.1	19.4	17.3	16.1	11.4	10.2	22.6	22.1
アイゾット衝撃値 (Vノチ) (J/m)	67	54	45	42	34	32	80	71
水素ガス透過性 ($\text{cm}^3/\text{m}^2 \cdot \text{atm} \cdot 24\text{h}$)	28	27	25	23	32	28	18	15
水蒸気透過性 ($\text{cm}^3/\text{m}^2 \cdot \text{atm} \cdot 24\text{h}$)	0.29	0.28	0.25	0.24	0.33	0.27	0.19	0.15
数平均繊維長/平均繊維径 (μm)	71	70	71	72	71	70	73	71
(成形品)	42	42	43	42	41	42	43	41
体積固有抵抗 ($\Omega \cdot \text{cm}$)	0.2	0.1	0.09	0.08	0.08	0.07	0.03	0.05

*1 PPS : ポリフェニレンスルフィド樹脂、LCP : 液晶ポリエステル樹脂

[0067] The constituent of this invention excels examples 1-8 in a mechanical property, gas barrier property, and conductivity, and it turns out that it is the material in which injection molding is possible.

[0068] It turns out that a mechanical property will fall a little if the loadings of a carbon fiber are fixed to 30wt(s)% and the quantity of the amount of carbon nanotubes is increased from examples 1-4, and conductivity goes up a little.

[0069] Although it excels in a mechanical property, gas barrier property, and conductivity and the material in which injection molding is possible is obtained from examples 5-7 even if it changes the loadings of a carbon fiber and a carbon nanotube a lot, even if it blends a carbon nanotube so much, conductive large improvement is not found, but the inclination for a mechanical property to fall is seen.

[0070] An example 8 shows that excel in a mechanical property, gas barrier property, and conductivity, and the material in which injection molding is possible is obtained, even if it changes a matrix resin.

[0071] The resin constituent was manufactured like the example 1 except carrying out the rate of a compounding ratio of [examples 1-2 of comparison] each component, as shown in Table 2. The rate of a compounding ratio and a characterization result are shown in Table 2.

[0072]

[Table 2]

表2

比較例	比較例1	比較例2	比較例3	比較例4
炭素繊維 (wt%)	30	—	黒鉛 76.8	ビッチ系炭素繊維 5
カーボンナノチューブ (wt%)	—	3	硬化剤 4	黒鉛 25 膨張性黒鉛 50
熱可塑性樹脂 *1 (種類)	PPS	PPS	エポキシ樹脂	LCP
(wt%)	70	97	19.2	20
引張強度 (MPa)	223	85	50	20
引張破断伸度 (%)	2.5	13.2	1.3	0.9
曲げ強度 (MPa)	267	142	111	27
曲げ弾性率 (GPa)	20.2	3.7	4.1	2.1
アイゾット衝撃値 (Vノット) (J/m)	68	45	44	4.1
水素ガス透過性 (cm ³ /m ² ・atm・24h)	28	46	53	52
水蒸気透過性 (cm ³ /m ² ・atm・24h)	0.28	0.48	0.88	0.86
数平均繊維長/平均繊維径 (μm)	70	—	—	—
(成形品)	42	—	—	—
体積固有抵抗 (Ω・cm)	149	1 × 10 ⁵	0.09	0.07

*1 PPS : ポリフェニレンスルフィド樹脂、LCP : 液晶ポリエステル樹脂

[0073] The examples 1-2 of comparison show that the material with which are satisfied of a mechanical property, gas barrier property, conductivity, and all the injection-molding nature is not obtained, when either a carbon nanotube or a carbon fiber is missing.

[0074] Continuation injection molding was performed using the metal mold of the configuration shown in drawing 1 using the constituent manufactured in the [example 9] example 3 in resin temperature [of 320 degrees C], 150 degrees-C [of die temperatures], and cooldown-delay 20 seconds. Even if it exceeded 1,000 shots of continuation, the faulty mold goods which are not were stabilized and were obtained. Drawing 1 is the plate which made the passage of hydrogen gas, oxygen gas, or air form in fuel cell separators, and is 3mm in size 66mmx84mm and thickness. The plate concerned is equipped with gas-passageway entrance A, the gas-passageway outlet B, and gas-passageway C. Moreover, the width of face of a gas passageway is 2.5mm, and the depth is 2mm.

[0075] The particle size of 110 micrometers or more and a graphite powder 160 micrometers or less are mixed by the ratio of the 20:80 weight sections as the end of the [example 3 of comparison] carbon powder to an epoxy resin (the product made from oil-ized shell epoxy "an Epicoat" 807), and, subsequently it is a curing agent (the 4 sections added and mixed degassing of the product IBMI 12 made from oil-ized shell epoxy "an epicure" was carried out.). Subsequently, using this material, press forming was carried out for 45 minutes by the temperature of 150 degrees C, and pressure 10MPa, and various evaluation test pieces were created. Furthermore, physical properties were evaluated after carrying out a cure at 150 degrees C after fabrication for 6 hours. A result is shown in Table 2.

[0076] Although it excels in conductivity as shown in Table 2, it turns out that it is inferior to a mechanical property.

[0077] the monotonous press of the configuration shown in drawing 1 using the material prepared in the example 3 of the [example 4 of comparison] comparison -- press forming was continuously performed using metal mold in the temperature of 150 degrees C, pressure 10MPa, and press time 45 minutes Breakage etc. occurred at the time of non-filling and mold release, and continuation press forming exceeding ten shots was not completed.

[0078] Melting extrusion of an expansibility graphite (expansibility graphite TEG[by Sumikin Chemical Co., Ltd.] 80 LTE-U), carbon black (KETCHIEN black EC by the KETCHIEN black international company), and the pitch based carbon fiber (Mitsubishi Chemical diamond lead K223 QG) was blended and carried out to the liquid crystal polyester resin manufactured in the example 2 of the [example 5 of comparison] reference at a rate shown in Table 2. It has not fabricated, although

injection molding was tried using the obtained pellet. Moreover, melting press forming was performed by the temperature of 340 degrees C, and pressure 10MPa, various evaluation test pieces were created, and physical properties were evaluated. A result is shown in Table 2. Although it excels in conductivity as shown in Table 2, it turns out that it is inferior to a mechanical property.

[0079]

[Effect of the Invention] By using this invention, the resin constituent and mold goods which are excellent in the moldability which was not obtained and have advanced conductivity can be obtained by the conventional method. Moreover, the fuel cell separator superior to the resin constituent of this invention can be obtained.

[Translation done.]

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PRIOR ART

[Description of the Prior Art] Since a carbon-dioxide-gas discharge problem is coped with in recent years, the method of supplying energy by the method which does not depend the energy acquired from petroleum resources on petroleum resources is examined. A fuel cell is in one of them, and development is briskly performed aiming at spread to the power supply for fuel cell vehicles, or a power supply for home use.

[0003] Each cell of a fuel cell is formed from the separator which supplies a gas passageway and a cooling water way while it contacts one pair of electrodes and the electrode which sandwich the electrolyte which manages a reaction, and an electrolyte and performs current collection from an electrode.

[0004] Among these, since it has a current collection function from an electrode, advanced conductivity is required for a separator. Moreover, in order to form fuel gas passage and a circulating-water-flow way in both sides, gas barrier property, intensity, a corrosion resistance, etc. are required. Furthermore, it is necessary to form a gas passageway and a circulating-water-flow way, and the outstanding moldability is required.

[0005] It is the method (JP,10-125337,A) of pressurizing and producing with the metal mold of the last configuration where the gas passageway and the circulating-water-flow way were attached, after pressurizing an expanded-graphite particle and making a reserve sheet form as a conventional method. A lot of graphites are blended with the way (JP,2000-36309,A) a fluororesin or a graphite fluoride particle forms an eutectoid and the distributed noble-metals compound plating coat in a metal separator front face into a noble-metals matrix, and thermosetting resin like phenol resin, and the method (JP,10-334927,A) of fabricating by press forming, the method (JP,2000-67882,A) of filling up precisely with conductive resin material the opening section of the metallic material which has the detailed opening section, the method (JP,2000-17179,A) of blending graphite system carbon material with a liquid crystal polymer so much, etc. are devised.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the monotonous planar structure view used in the example 9 and the example 4 of comparison.

[Description of Notations]

A: Gas-passageway entrance

B: Gas-passageway outlet

C: Gas passageway

[Translation done.]

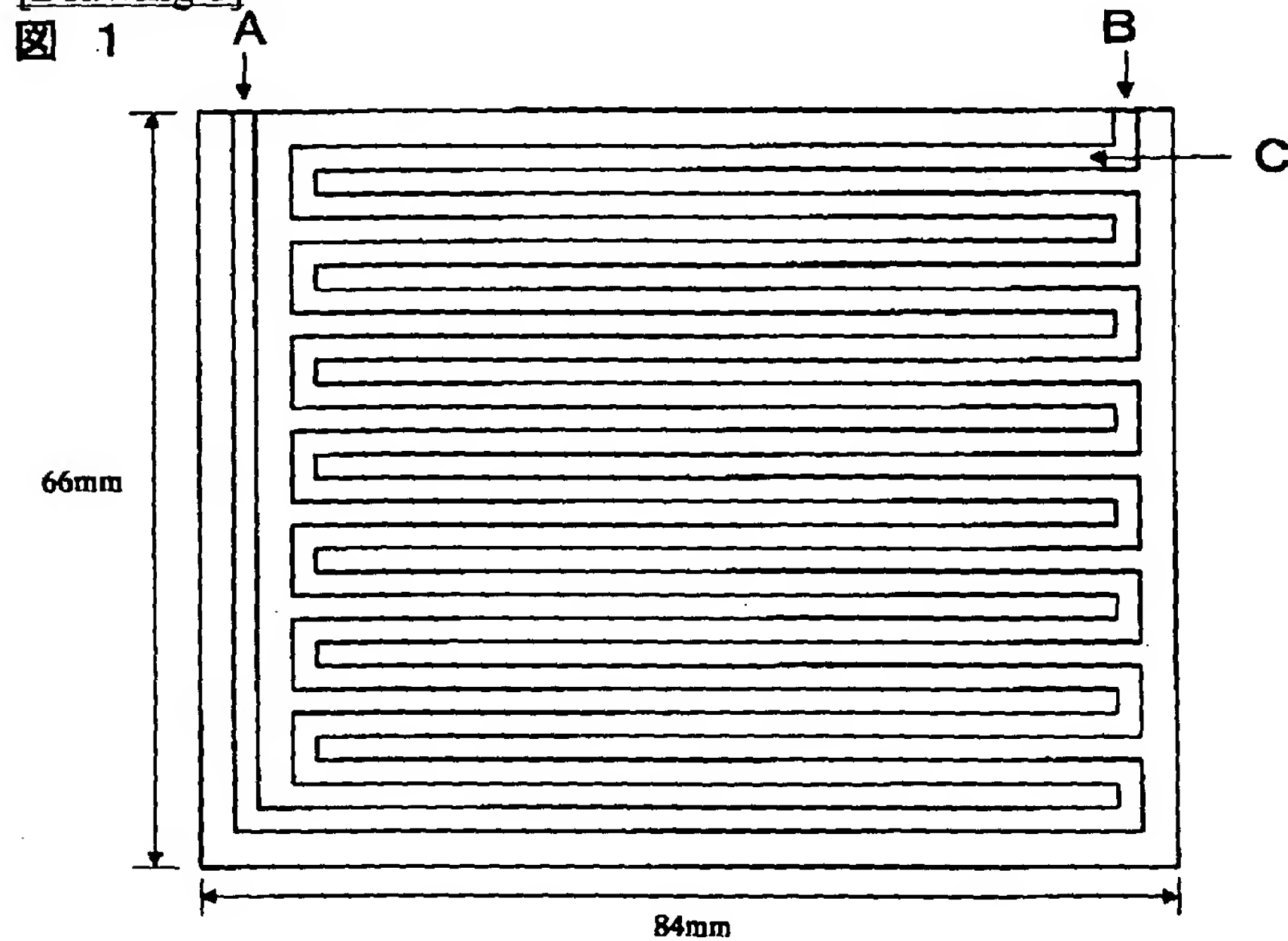
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DRAWINGS

[Drawing 1]



[Translation done.]

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(72)Inventor : OKITA SHIGERU
SUZUKI ATSUSHI

(54) THERMOPLASTIC RESIN COMPOSITION AND MOLDING

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a thermoplastic resin composition having excellent electroconductivity, gas barrier properties, strength, corrosion resistance and moldability, suitable for a separator for a fuel cell.

SOLUTION: This thermoplastic resin composition is obtained by compounding a thermoplastic resin with a carbon fiber and a carbon nanotube.

LEGAL STATUS

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